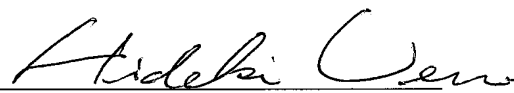


I, Hideki Ueno, residing at 17-5, Tsurumaki 3-chome, Setagaya-ku, Tokyo 154-0016, Japan, and working for ISP Corporation of 1-29, Akashi-cho, Chuo-ku, Tokyo 104-0044, Japan, fully conversant with the English and Japanese languages, do hereby certify that to the best of my knowledge and belief the following is a true translation of Japanese Patent Application No. 2003-208264 filed in the Japanese Patent Office on the 21st day of August, 2003 in respect of an application for Letters Patent.

Signed, this 27th day of October, 2009



Hideki Ueno

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[TITLE OF THE INVENTION] LOW FRICTION SLIDING MECHANISM, LOW
FRICTION AGENT COMPOSITION AND FRICTION REDUCING METHOD

[DOCUMENT TITLE] SCOPE OF CLAIM FOR PATENT

1. A low-friction sliding mechanism wherein an oxygen-containing organic compound (C) or a low-friction agent composition (D) is interposed between sliding surfaces of a DLC coated sliding member (A) and a sliding member (B), characterized in that the DLC coated sliding member (A) is formed by coating diamond-like carbon on

a base material;

the sliding member (B) is formed with at least one kind of material selected from a group consisting of a metal material, a non-metal material and a coated material obtained by coating a thin film on a surface of the metal material or the non-metal material;

the oxygen-containing organic compound (C) includes an oxygen in a molecule; and

the low-friction agent composition (D) contains the oxygen-containing organic compound (C).

2. The low-friction sliding mechanism according to claim 1, characterized in that the DLC coated sliding member (A) is coated with a-C diamond-like carbon that does not contain hydrogen.
3. The low-friction sliding mechanism according to claim 1 or 2, characterized in that, in the sliding member (B), the metal material is at least one kind of material selected from a group consisting of a ferrous material, an aluminum alloy material and a magnesium alloy-based material; and the coated material is coated with a thin film of at least one kind of material selected from a group consisting of DLC, TiN and CrN.
4. The low-friction sliding mechanism according to claim 3, characterized in that the coated DLC is made of a-C diamond-like carbon that does not contain hydrogen.
5. The low-friction sliding mechanism according to any one of claims 1 through 4, characterized in that the oxygen containing organic compound (C) is at least one kind selected from a group consisting of alcohols, carboxylic acids, esters, ethers, ketones, aldehydes, carbonates and derivatives thereof.
6. The low-friction sliding mechanism according to any one of claims 1 through 5, characterized in that the low-friction agent composition (D) contains a grease-like or a wax-like medium and the oxygen-containing organic compound (C) at the time of using the (D).
7. The low-friction sliding mechanism according to claim 6, characterized in that the medium is a lubricant.
8. A low-friction agent composition, characterized by an oxygen containing organic compound used in the low-friction sliding mechanism according to any of claims 1 to 7 and containing an oxygen in a molecule.
9. A method of friction reduction characterized in that, on sliding surfaces formed of a DLC coated sliding member (A) formed by coating diamond-like carbon and

a sliding member (B) that uses at least one kind of material selected from a group consisting of a metal material, a non-metal material and a coated material obtained by coating a thin film on a surface of the metal material or the non-metal material, one that contains at least one kind selected from a group consisting of an oxygen-containing organic compound (C) and a low-friction agent composition (D) to lubricate.

[DETAILED DESCRIPTION OF THE INVENTION]

[0001]

[TECHNICAL FIELD]

The present invention relates to a low-friction sliding mechanism, a low-friction agent composition, and a method of friction reduction. In more detail, the present invention relates to a low-friction sliding mechanism a low-friction agent composition, and a method of friction reduction that can improve the friction characteristics of various sliding surfaces in, for instance, internal combustion engines and drive system transmission units.

[0002]

[BACKGROUND ART]

Earth scale environment problems such as global warming and destruction of the ozone layer are largely highlighted. In particular, the CO₂ reduction that is said to largely affect on the warming of the earth as a whole is gathering attention on how to decide the regulation value in each country. As to the CO₂ reduction, the reduction of the energy loss due to the friction loss of machines and devices, in particular, the reduction of the fuel cost of automobiles is a large problem. Accordingly, the roles of sliding materials and lubricants are very important in this connection. A role of the sliding material is to be excellent in the wear resistance and develop excellent wear resistance to sections of which friction and wear environment are severe among the sliding sections of an engine. Recently, various kinds of hard thin film materials are being forwarded in applications. The friction coefficients of general DLC materials are lower in air and in the absence of lubricating oil than that of wear resistant hard coating materials such as TiN and CrN; accordingly, these are expected as low friction sliding materials.

[0003] Furthermore, as an energy saving measure in the lubricating oil, for instance, as a fuel efficiency measure of an engine, 1) reduction, caused by lowering the viscosity, of the viscous resistance in a hydrodynamic lubrication region and the agitation resistance in

an engine and 2) reduction, owing to blending of an optimum friction modifier and various kinds of additives, of the friction loss under mixed lubrication and boundary lubrication region are proposed. Many researches have been conducted mainly of organic Mo compounds such as MoDTC and MoDTP as the friction modifier. In a sliding surface made of an existing steel material, a lubricating oil composition in which an organic Mo compound that exhibits the excellent low friction coefficient at an early stage of use is advantageously applied.

[0004] On the other hand, a general DLC material excellent in the low friction characteristics in air is reported to be, in the presence of the lubricating oil, low in the friction reduction effect (for instance, non-patent literature 1). Furthermore, it has been found that even when a lubricating oil composition containing an organic molybdenum compound is applied to the sliding material the friction reduction effect cannot be sufficiently exerted (for instance, non-patent literature 2).

[0005]

[Non-patent literature 1] Kano et al., Proceedings of Japanese Tribology Society, p11 to 12, May, 1999 (Tokyo)

[0006]

[Non-patent literature 2] Kano et al., Proceeding of World Tribology Congress Sep., 2001, p. 342, (Vienna)

[PROBLEM TO BE SOLVED BY THE INVENTION]

[0007] The invention is achieved considering such problems that existing technologies have; and intends to provide a low-friction sliding mechanism, a low-friction agent composition and a friction reduction method that can exert very excellent low friction characteristics to a sliding surface present under various applications, and, in particular, that have more excellent low friction characteristics than that of a combination of the existing steel material and the organic Mo compound.

[0008]

[MEANS FOR THE SOLUTION TO THE PROBLEM]

The inventors, after studying hard to overcome the above problems, found that a combination of sliding members at least one of which is a DLC material and a specific compound shows the friction characteristics very excellent to an extent that cannot be realized from an existing lubrication theory, and thereby come to completion of the invention.

[0009]

[EMBODIMENTS OF THE INVENTION]

In what follows, a low-friction sliding mechanism and a low-friction agent composition according to the invention will be more detailed. In the specification, "%" denotes a mass percentage unless otherwise stated.

[0010] Such a low-friction sliding mechanism is formed by including a low-friction agent composition between sliding surfaces that the sliding members form when a DLC coated sliding member (A) and a sliding member (B) are slid. As the low-friction agent composition, one that contains at least one kind selected from a group consisting of an oxygen containing organic compound (C) and an aliphatic amine compound (D) is used. Thereby, the DLC coated sliding member and the sliding member slide each other at the extremely lower friction than ever.

[0011] Here, a DLC (Diamond-Like Carbon) material used in the DLC coated sliding member (A) is an amorphous mainly made of carbon element, and a bonding state between carbons is made of both of a diamond structure (SP^3 bond) and a graphite bond (SP^2 bond). Specifically, a-C (amorphous carbon) entirely made of carbon element, a-C: H (hydrogen amorphous carbon) containing hydrogen and MeC partially containing a metal element such as titanium (Ti) or molybdenum (Mo) can be cited. In the low-friction sliding mechanism according to the invention, from a viewpoint of expressing a large friction reduction effect, the DLC material is preferably made of an a-C material that does not contain hydrogen. Furthermore, as a base material that is used in the DLC coated sliding member (A), for instance, carburized steel, hardened steel and non-ferrous metals such as aluminum can be used.

[0012] Furthermore, a constituent material of the sliding member (B) is not particularly restricted. Specifically, metal materials such as ferrous materials, aluminum base materials, magnesium base materials and titanium base materials can be cited. In particular, the ferrous materials, aluminum base materials and magnesium base materials, being readily applicable to sliding sections of existing machines and devices and able to widely contribute to an energy saving measure in various fields, are preferable. Still furthermore, as the constituent materials of the sliding member (B), non-metal materials such as resins, plastics and carbon can be used as well. Furthermore, materials obtained by applying various kinds of thin film coating on the metal materials and nonmetal materials are useful as well.

[0013] The ferrous materials are not particularly restricted. Not only high purity

iron, but also various kinds of ferrous alloys (nickel, copper, zinc, chromium, cobalt, molybdenum, lead, silicon or titanium, and ones obtained by arbitrarily combining these) can be used. Specifically, for instance, carburized steel SCM420 or SCr420 (JIS) can be cited. Furthermore, the aluminum base materials are not particularly restricted. Not only high purity aluminum but also various kinds of aluminum base alloys can be used. Specifically, for instance, a hypoeutectic aluminum alloy containing 4 to 20% silicon (Si) and 1.0 to 5.0% copper or a hypereutectic aluminum alloy can be desirably used. As preferable example of the aluminum alloy, for instance, AC2A, AC8A, ADC12 and ADC14 (JIS) can be cited.

[0014] Among the sliding members (B), the metal materials provided with various kinds of coating are not particularly restricted. Specifically, various kinds of metal materials such as metal base materials obtained by applying a thin film of TiN, CrN or the DLC material on a surface of the ferrous materials, aluminum base materials, magnesium base materials or titanium base materials can be cited. Among these, the metal material coated with the DLC material is preferable. Furthermore, the DLC material is preferably the a-C diamond-like carbon that does not contain hydrogen.

[0015] Furthermore, from the sliding stability, the surface roughness Ra of each of the DLC coated sliding member (A) and the sliding member (B) (for instance, metal materials or the metal materials coated with various kinds of thin film) is 0.1 μm or less and preferably 0.08 μm or less. When it exceeds 0.1 μm , local scaffing is caused to result in a large increase in the friction coefficient in some cases.

[0016] Still furthermore, in the DLC coated sliding member (A), it is preferable that the surface hardness thereof is in the range of Hv1000 to 3500 in the micro-Vickers hardness (10 g load) and a DLC film thickness is in the range of 0.3 to 2.0 μm . When the surface hardness and the thickness of the DLC coated sliding member (A) are outside of the abovementioned ranges to be less than Hv1000 in the surface hardness and less than 0.3 μm in the thickness, the DLC coated sliding member becomes easily worn, by contrast, when they exceed Hv3500 and 2.0 μm , respectively, it becomes easily peeled, and, when the surface hardness of the ferrous member is outside thereof to be less than HRC45, under high pressure, in some cases, it becomes easier to buckle and peel. In the case of a ferrous material being used in the sliding member (B), the surface hardness thereof is preferably in the range of HRC45 to 60 in Rockwell

hardness C-scale. In this case, even under a high surface pressure sliding condition such as substantially 700 MPa like in a cam follower member, the durability of the film can be effectively maintained. Furthermore, in the case of an aluminum base material being used in the sliding member (B), the surface hardness thereof is preferably in the range of HB80 to 130 in Brinell hardness. When the surface hardness of the aluminum base material is outside of the above range to be less than HB80, the aluminum base material becomes easily worn. Still furthermore, in the case of a thin film-coated metal material being used in the sliding member (B), in particular, in the case of the DLC-coated metal material being used, it is preferable that the surface hardness thereof is in the range of Hv1000 to 3500 in the micro-Vickers hardness (10 g load) and a DLC film thickness is in the range of 0.3 to 2.0 μm . When the surface hardness and the thickness thereof are outside of the above-mentioned ranges to be less than abrasion is easily caused, by contrast, when they exceed Hv3500 and 2.0 μm , in some cases, peeling is easily caused.

[0017] Sliding surfaces made of the DLC coated sliding member (A) and the sliding member (B), as long as these are sliding surfaces where two sliding surfaces come into contact through a low-friction agent composition, can be used without particular restrictions. For instance, sliding sections of internal combustion engines such as 4-cycle engines and 2-cycle engines (for instance, a valve operating system, a piston, a piston ring, a piston skirt, a cylinder liner, a con-rod, a crank shaft, a bearing, a roller bearing, metal, a gear, a chain, a belt, an oil pump and the like), sliding sections of a driving system transmission mechanism (for instance, gear) and a hard disc drive, and other various sliding surfaces where the friction conditions are severe and the low friction properties are demanded are targeted. In the sliding surfaces, when only at least one of the sliding members is DLC-coated and at least one kind selected from the oxygen-containing organic compound and aliphatic amine compound is supplied, more than ever lubricant and extremely excellent low friction characteristics can be effectively obtained. For instance, as preferable embodiments in a valve operating system of an internal combustion engine, sliding surfaces made of a disc-like shim and a lifter crown surface where the DLC is coated on a substrate of a steel material and a cam lobe that uses a material involving low-alloy chilled iron, carburized steel or tempered carbon steel, and arbitrary combinations thereof can be cited.

[0018] On the other hand, the aforementioned oxygen-containing organic

compound (C) in the low-friction sliding mechanism of the invention is not particularly restricted and can be any organic compound containing oxygen in the molecule. For example, it may be an oxygen-containing organic compound constituted of carbon, hydrogen and oxygen. The oxygen-containing organic compound may contain another element such as nitrogen, sulfur, halogen (fluorine, chlorine etc.), phosphorus, boron, a metal, or the like. Particularly in view of further reducing the friction in the sliding surfaces of one sliding member (DLC coated sliding member) (A) and the other sliding member (B), there is preferred an oxygen-containing organic compound constituted of carbon, hydrogen and oxygen and containing a hydroxyl group, or a derivative thereof, in which it is more preferred that two or more hydroxyl groups are contained. Also for the same reason, there is more preferred an oxygen-containing organic compound with a low sulfur content or free from sulfur. Also a "derivative" used herein is not particularly restricted and can be, for example, a compound obtained by reacting an oxygen-containing organic compound constituted of carbon, hydrogen and oxygen with a nitrogen-containing compound, a phosphorus-containing compound, sulfur or a sulfur-containing compound, a boron-containing compound, a halogen element or a halogen-containing compound, a metal element, a metal-containing compound (organic or inorganic), or the like.

[0019] The aforementioned oxygen-containing organic compound (C) can specifically be a compound containing a hydroxyl group, a carboxyl group, a carbonyl group, an ester bond or an ether bond (two or more kinds of such group or bond may also be contained), preferably contains one or more kinds of group or bond selected from a hydroxyl group, a carboxyl group, a carbonyl group, and an ester bond, more preferably is an oxygen-containing organic compound containing one or more kinds of group or bond selected from a hydroxyl group, a carboxyl group and an ester group, further preferably is an oxygen-containing organic compound containing one or more kinds of group selected from a hydroxyl group and a carboxyl group, and particularly preferably an oxygen-containing organic compound containing one or more hydroxyl groups.

[0020] More specific examples of the oxygen-containing organic compound include alcohols (I), carboxylic acids (II), esters (III), ethers (IV), ketones (V), aldehydes (VI), carbonates (these compounds may further contain one or more kinds of group or bond selected from a hydroxyl group, a carboxyl group, a carbonyl group, an ester bond and an ether bond) (VII), derivatives thereof and

an arbitrary mixture thereof.

[0021] The above-mentioned alcohols (I) are oxygen-containing organic compounds represented by the following general formula (1)

[0022]

$R-(OH)_n$ (1)

[0023]

,for example, a compound containing one or more hydroxyl groups.

[0024] Examples of alcohols (I) are mentioned below:

Monoalcohols (I-1);

Dialcohols (I-2);

Tri and higher polyalcohols (I-3); and

Alkyl oxydide additive of the above three kinds of alcohols (I-4)

Mixtures of one or more selected from the above four kinds of alcohols (I-5).

[0025] Monoalcohols (I-1) have one hydroxyl group in the molecule, including, for example, monohydric alkyl alcohols having from 1 to 40 carbon atoms (in which the alkyl group may be linear or branched) such as methanol, ethanol, propanol (1-propanol, 2-propanol), butanol (1-butanol, 2-butanol, 2-methyl-1-propanol, 2-methyl-2-propanol), pentanol (1-pentanol, 2-pentanol, 3-pentanol, 2-methyl-1-butanol, 3-methyl-1-butanol, 3-methyl-2-butanol, 2-methyl-2-butanol, 2,2-dimethyl-1-propanol), hexanol (1-hexanol, 2-hexanol, 3-hexanol, 2-methyl-1-pentanol, 2-methyl-2-pentanol, 2-methyl-3-pentanol, 3-methyl-1-pentanol, 3-methyl-2-pentanol, 3-methyl-3-pentanol, 4-methyl-1-pentanol, 4-methyl-2-pentanol, 2,3-dimethyl-1-butanol, 2,3-dimethyl-2-butanol, 3,3-dimethyl-1-butanol, 3,3-dimethyl-2-butanol, 2-ethyl-1-butanol, 2,2-dimethylbutanol), heptanol (1-heptanol, 2-heptanol, 3-heptanol, 2-methyl-1-hexanol, 2-methyl-2-hexanol, 2-methyl-3-hexanol, 5-methyl-2-hexanol, 3-ethyl-3-pentanol, 2,2-dimethyl-3-pentanol, 2,3-dimethyl-3-pentanol, 2,4-dimethyl-3-pentanol, 4,4-dimethyl-2-pentanol, 3-methyl-1-hexanol, 4-methyl-1-hexanol, 5-methyl-1-hexanol, 2-ethylpentanol), octanol (1-octanol, 2-octanol, 3-octanol, 4-methyl-3-heptanol, 6-methyl-2-heptanol, 2-ethyl-1-hexanol, 2-propyl-1-pentanol, 2,4,4-trimethyl-1-pentanol, 3,5-dimethyl-1-hexanol, 2-methyl-1-heptanol, 2,2-dimethyl-1-hexanol), nonanol (1-nonanol, 2-nonanol, 3,5,5-trimethyl-1-hexanol, 2,6-dimethyl-4-heptanol, 3-ethyl-2,2-dimethyl-3-pentanol, 5-methyloctanol, etc.), decanol (1-decanol,

2-decanol, 4-decanol, 3,7-dimethyl-1-octanol, 2,4,6-trimethylheptanol, etc.), undecanol, dodecanol, tridecanol, tetradecanol, pentadecanol, hexadecanol, heptadecanol, octadecanol (stearyl alcohol, etc.), nonadecanol, eicosanol, heneicosanol, tricosanol, tetracosanol; monohydric alkenyl alcohols having from 2 to 40 carbon atoms (in which the alkenyl group may be linear or branched and the double bond may be in any desired position) such as ethenol, propenol, butenol, hexenol, octenol, decenol, dodecenol, octadecenol (oleyl alcohol, etc.); monohydric (alkyl)cycloalkylalcoholshavingfrom 3 to 40 carbon atoms (in which the alkyl group may be linear or branched, and the alkyl group and the hydroxyl group may be in any desired position) such as cyclopentanol, cyclohexanol, cycloheptanol, cyclooctanol, methylcyclopentanol, methylcyclohexanol, dimethylcyclohexanol, ethylcyclohexanol, propylcyclohexanol, butylcyclohexanol, dimethylcyclohexanol, cyclopentylmethanol, cyclohexylethanol (1-cyclohexylethanol, 2-cyclohexylethanol, etc.), cyclohexylethanol, cyclohexylpropanol (3-cyclohexylpropanol, etc.), cyclohexylbutanol (4-cyclohexylbutanol, etc.)

butylcyclohexanol, 3,3,5,5-tetramethylcyclohexanol; (alkyl) aryl alcohols (in which the alkyl group may be linear or branched, and the alkyl group and the hydroxyl group may be in any desired position) such as phenyl alcohol, methylphenyl alcohol (o-cresol, m-cresol, p-cresol), creosol, ethylphenyl alcohol, propylphenyl alcohol, butylphenyl alcohol, butylmethylphenyl alcohol (3-methyl-6-tert-butylphenyl alcohol, etc.), dimethylphenyl alcohol, diethylphenyl alcohol, dibutylphenyl alcohol (2,6-di-tert-butylphenyl alcohol, 2,4-di-tert-butylphenyl alcohol, etc.), dibutylmethylphenyl alcohol (2,6-di-tert-butyl-4-methylphenyl alcohol, etc.), dibutylethylphenyl alcohol (2,6-di-tert-butyl-4-ethylphenyl alcohol, etc.), tributylphenyl alcohol (2,4,6-tri-tert-butylphenyl alcohol, etc.), naphthol (a-naphthol, b-naphthol, etc.), dibutylnaphthol (2,4-di-tert-butyl-a-naphthol, etc.); 6-(4-^L hydroxy-3,5-di-tert-butylanilino)-2,4-bis(n-octylthio)-1,3,5-triazine, and their mixtures.

[0026] Of those, more preferred are linear or branched alkyl or alkenyl alcohols having from 12 to 18 carbon atoms such as oleyl alcohol and stearyl alcohol, in that they may more effectively lower the friction of the sliding surfaces formed of the DLC coated sliding member (A) and any other sliding member (B) and that they are poorly volatile and therefore may exhibit their friction-^L reducing effect even at high temperature conditions (for example, sliding condition in an internal

combustion engine).

[0027] Dialcohols (I-2) are concretely those having two hydroxyl groups in the molecule, including, for example, alkyl or alkenyldiols having from 2 to 40 carbon atoms (in which the alkyl or alkenyl group may be linear or branched, the double bond of the alkenyl group may be in any desired position, and the hydroxyl group may also be in any desired position) such as ethylene glycol, diethylene glycol, polyethylene glycol, propylene glycol, dipropylene glycol, polypropylene glycol, neopentyl glycol, 1,3-propanediol, 1,4-butanediol, 1,2-butanediol, 2-methyl-1,3-propanediol, 1,5-pentanediol, 1,6-hexanediol, 2-ethyl-2-methyl-1,3-propanediol, 2-methyl-2,4-pentanediol, 1,7-heptanediol, 2-methyl-2-propyl-1,3-propanediol, 2,2-diethyl-1,3-propanediol, 1,8-octanediol, 1,9-nonanediol, 2-butyl-2-ethyl-1,3-propanediol, 1,10-decanediol, 1,11-undecanediol, 1,12-dodecanediol, 1,13-tridecanediol, 1,14-tetradecanediol, 1,15-heptadecanediol, 1,16-hexadecanediol, 1,17-heptadecanediol, 1,18-octadecanediol, 1,19-nonadecanediol, 1,20-eicosadecanediol; (alkyl) cycloalkanediols

(in which the alkyl group may be linear or branched, and the alkyl group and the hydroxyl group may be in any desired position) such as cyclohexanediol, methylcyclohexanediol; dihydric (alkyl) aryl alcohols having from 2 to 40 carbon atoms (in which the alkyl group may be linear or branched, and the alkyl group and the hydroxyl group may be in any desired position) such as benzenediol (catechol, etc.), methylbenzenediol, ethylbenzenediol, butylbenzenediol (p-tert-butylcatechol, etc.) dibutylbenzenediol (4,6-di-tert-butylresorcinol, etc.), 4,4'-thiobis(3-methyl-6-tert-butylphenol), 4,4'-butylidenebis(3-methyl-6-tert-butylphenol), 2,2'-methylenebis(4-methyl-6-tert-butylphenol), 2,2'-thiobis(4,6-di-tert-butylresorcinol), 2,2'-methylenebis(4-ethyl-6-tert-butylphenol), 4,4'-methylenebis^L(2,6-di-tert-butylphenol), 2,2'-(3,5-ditert-butylhydroxy)propane, 4,4'-cyclohexylidenebis(2,6-di-tert-butylphenol); p-tert-butylphenol/formaldehyde condensate,

p-tert-butylphenol/acetaldehyde condensate; and their mixtures.

[0028] Of those, preferred are ethylene glycol, propylene glycol, neopentyl glycol, 1,4-butanediol, 1,5-pentanediol,

neopentyl glycol, 1,6-hexanediol, 2-methyl-2,4-pentanediol, 2-ethyl-2-methyl-1,3-propanediol, 1,7-heptanediol, 1,8-octanediol, 1,9-nonanediol, 1,10-decanediol, 1,11-undecanediol and 1,12-dodecanediol, in that they may more effectively lower the friction at the sliding surfaces of the DLC coated sliding member (A) and of any sliding member (B). In addition, high-molecular-weight hindered alcohols having a molecular weight of at least 300, preferably at least 400 such as 2,6-di-tert-butyl-4-(3,5-di-tert-butyl-4-hydroxybenzyl) phenyl alcohol are also preferred in that they are hardly volatile even at high temperatures (for example, under sliding condition in internal-combustion engines) and are highly resistant to heat, and they can well exhibit their friction-reducing effect and can impart excellent anti oxidation stability to lubricating oil.

[0029] Tri- and higher polyalcohols (I-3) are concretely those having three or more hydroxyl groups. In general, tri- to deca-alcohols, preferably tri- to hexa-alcohols are used. Examples of these components are trimethylolalkanes such as glycerin, trimethylolethane, trimethylolpropane, trimethylolbutane; as well as erythritol, pentaerythritol, 1,2,4-butanetriol,

1,3,5-pentanetriol, 1,2,6-hexanetriol, 1,2,3,4-butanetetrol, sorbitol, adonitol, arabitol, xylitol, mannitol; and their polymers or condensates (e.g., glycerindimers to octamers such as diglycerin, triglycerin, tetraglycerin; trimethylolpropane dimmers to octamers such as ditrimethylolpropane; pentaerythritol dimers to tetramers such as dipentaerythritol; sorbitan; condensates such as sorbitol/glycerin condensate (including intramolecular condensates, intermolecular condensates, and self-condensates)).

[0030] Saccharides such as xylose, arabitol, ribose, rhamnose, glucose, fructose, galactose, mannose, sorbose, cellobiose, mannose, isomaltose, trehalose and sucrose are also usable.

[0031] Of those, more preferred are tri to hexa-alcohols such as glycerin, trimethylolalkanes (e.g., trimethylolethane, trimethylolpropane, trimethylolbutane), pentaerythritol, 1,2,4-butanetriol, 1,3,5-pentanetriol, 1,2,6-hexanetriol, 1,2,3,4-butanetetrol, sorbitol, sorbitan, sorbitol/^L glycerin condensate, adonitol, arabitol, xylitol, mannitol, and their mixtures; and even more preferred are glycerin, trimethylolethane, trimethylolpropane, pentaerythritol, sorbitan and their mixtures. Especially preferred are polyalcohols having an oxygen content of at least 20 %, preferably at least 30 %, more preferably at least 40 %. Polyalcohols that are higher than

hexa-alcohols will too much increase the viscosity.

[0032] Further, an alkylene oxide additive (1-4) is an alkylene oxide additive of alcohols (1-1 to 3), and specially alkylene oxide of C 2 to 6, preferably C 2 to 4, the polymer or copolymer is added to the alcohols and hydroxyl group of alcohols are haidorokarubi-etherified or haidorokarubi-esterified. Alkylene oxide of C 2 to 6, ethylene oxide, propylene oxide, 1,2-epoxy butane (α -butylene oxide), 2,3 epoxy butane (β -butylene oxide), 1, 2-epoxy-1-methyl propane, 1-2-epoxy heputane, 1, 2 epoxy hexane and the like are exemplified. Among them, from a viewpoint of excellent low friction properties, ethylene oxide, propane oxide and butylene oxide are preferable, and ethylene oxide and propane oxide are more preferable.

[0033] In a case of using two or more kinds of alkylene oxide, there is no limit to polymer form of oxy alkylene group, and random copolymer or block copolymer may be allowed. Upon adding alkylene oxide to poly alcohol having 2 to 6 pieces of hydroxyl group, it may add to all hydroxyl group or a part of hydroxyl group.

[0034] The above-mentioned carboxylic acids (II) are compounds represented by the following general formula (2):

[0035]



[0036]

, for example, a compound containing one or more carboxyl groups.

[0037] Examples of carboxylic acids (II) are mentioned below:

Aliphatic monocarboxylic acids (fatty acids)(II-1);

Aliphatic polycarboxylic acids(II-2);

Carbon-cyclic carboxylic acids (II-3);

Heterocyclic carboxylic acids (II-4); and

Mixtures of two or more selected from the above four kinds of carboxylic acids(II-5).

[0038] Aliphatic monocarboxylic acids (fatty acids) (II-1) are concretely those having one carboxyl group in the molecule, including, for example, saturated aliphatic monocarboxylic acids having from 1 to 40 carbon atoms (in which the saturated aliphatic structure may be linear or branched) such as methanoic acid, ethanoic acid (acetic acid), propanoic acid (propionic acid), butanoic acid (butyric acid, isobutyric acid, etc.), pentanoic acid (valeric acid, isovaleric acid, pivalic acid, etc.), hexanoic acid (caproic acid, etc.), heptanoic acid, octanoic

acid (caprylic acid, etc.), nonanoic acid (pelargonic acid, etc.), decanoic acid, undecanoic acid, dodecanoic acid (lauric acid, etc.), tridecanoic acid, tetradecanoic acid (myristic acid, etc.), pentadecanoic acid, hexadecanoic acid (palmitic acid, etc.), heptadecanoic acid, octadecanoic acid (stearic acid, etc.), nonadecanoic acid, eicosanoic acid, heneicosanoic acid, docosanoic acid, tricosanoic acid, tetracosanoic acid, pentacosanoic acid, hexacosanoic acid, heptacosanoic acid, octacosanoic acid, nonacosanoic acid, triacontanoic acid; and unsaturated aliphatic monocarboxylic acids having from 1 to 40 carbon atoms (in which the unsaturated aliphatic structure maybe linear or branched, and the unsaturated bond may be in any desired position) such as propenoic acid (acrylic acid, etc.), propynoic acid (propiolic acid, etc.), butenoic acid (methacrylic acid, crotonic acid, isocrotonic acid, etc.), pentenoic acid, hexenoic acid, heptenoic acid, octenoic acid, nonenoic acid, decenoic acid, undecenoic acid, dodecenoic acid, tridecenoic acid, tetradecenoic acid, pentadecenoic acid, hexadecenoic acid, heptadecenoic acid, octadecenoic acid (oleic acid, etc.), nonadecenoic acid, eicosenoic acid, heneicosenoic acid, docosenoic acid, tricosenoic acid, tetracosenoic acid, pentacosenoic acid, hexacosenoic acid, heptacosenoic acid, octacosenoic acid, nonacosenoic acid, triacontenoic acid.

[0039] Aliphatic polycarboxylic acids (II-2) include saturated or unsaturated aliphatic dicarboxylic acids having from 2 to 40 carbon atoms (in which the saturated aliphatic or unsaturated aliphatic structure maybe linear or branched, and the unsaturated bond may be in any desired position) such as ethane-diacid (oxalic acid), propane-diacid (malonic acid, etc.), butane-diacid (succinic acid, methylmalonic acid, etc.), pentane-diacid (glutaric acid, ethylmalonic acid, etc.), hexane-diacid (adipic acid, etc.), heptane-diacid (pimelic acid, etc.), octane-diacid (suberic acid, etc.), nonane-diacid (azelaic acid, etc.), decane-diacid (sebacic acid, etc.), propene-diacid, butene-diacid (maleic acid, fumaric acid, etc.), pentene-diacid (citraconic acid, mesaconic acid, etc.), hexene-diacid, heptene-diacid, octene-diacid, nonene-diacid, decene-diacid; saturated or unsaturated tricarboxylic acids (in which the saturated aliphatic or unsaturated aliphatic structure may be linear or branched, and the unsaturated bond may be in any desired position) such as propanetricarboxylic acid, butane-tricarboxylic acid, pentane-tricarboxylic acid, hexane-tricarboxylic acid, heptane-tricarboxylic acid, octane-tricarboxylic acid, nonane-tricarboxylic acid, decane-tricarboxylic acid; and saturated or unsaturated tetracarboxylic acids (in which the saturated aliphatic or unsaturated aliphatic structure may be

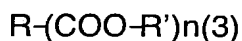
linear or branched, and the unsaturated bond may be in any desired position).

[0040] Carbon-cyclic carboxylic acids (II-3) are concretely those having one or more carboxyl groups in the carboncyclic molecule, including, for example, naphthene ring-having, mono, di, tri or tetracarboxylic acids having from 3 to 40 carbon atoms (in which the alkyl or alkenyl group, if any therein, maybe linear or branched, and the double bond, if any therein, may be in any desired position, and the number and the position of the substituents are not defined) such as cyclohexane-monocarboxylic acid, methylcyclohexane-monocarboxylic acid, ethylcyclohexane-monocarboxylic acid, propylcyclohexane-monocarboxylic acid, butylcyclohexane-monocarboxylic acid, pentylcycohexane-monocarboxylic acid, hexylcyclohexane-monocarboxylic acid, heptylcyclohexane-monocarboxylic acid, octylcyclohexane-monocarboxylic acid, cycloheptane-monocarboxylic acid, cyclooctane-monocarboxylic acid, trimethylcyclopentane-dicarboxylic acid (camphor acid, etc.); aromatic monocarboxylic acids having from 7 to 40 carbon atoms such as benzenecarboxylic acid (benzoic acid), methylbenzenecarboxylic acid (toluic acid, etc.), ethylbenzenecarboxylic acid, propylbenzenecarboxylic acid, benzenedicarboxylic acid (phthalic acid, isophthalic acid, terephthalic acid, etc.), benzenetricarboxylic acid^L (trimellitic acid, etc.), benzeneteracarboxylic acid (pyromellitic acid, etc.), naphthalenecarboxylic acid (naphthoic acid, etc.) ; mono, di, tri or tetracarboxylic acids having an aryl group with from 7 to 40 carbon atoms (in which the alkyl or alkenyl group, if any therein as a substituent, may be linear or branched and the double bound, if any therein, may be in any desired position, and the number and the position of the substituents are not defined) such as phenylpropanoic acid (hydroatropic acid), phenylpropenoic acid (atropic acid, cinnamic acid, etc.), salicylic acid, alkylsalicylic acid having one or more alkyl groups with from 1 to 30 carbon atoms.

[0041] Heterocyclic carboxylic acids (II-4) are concretely those having one or more carboxyl groups in the molecule, including, for example, those having from 5 to 40 carbon atoms such as furanecarboxylic acid, thiophenecarboxylic acid, pyridinecarboxylic acid (nicotinic acid, isonicotinic acid, etc.).

[0042] The above-mentioned esters (III) are compounds represented by the following general formula (3):

[0043]



[0044]

,for example, a compound containing one or more ester bonds.

[0045] Examples of esters (III) are mentioned below:

Esters of aliphatic monocarboxylic acids (fatty acids) (III-1);

Esters of aliphatic polycarboxylic acids (III-2);

Esters of carbon-cyclic carboxylic acids (III-3);

Esters of heterocyclic carboxylic acids (III-4);

Mixtures of any compounds selected from the above five kinds of esters (III-5) .

Esters of the above III-1 to III-5 may be complete esters in which the hydroxyl group and the carboxyl group are all esterified, or partial esters in which the hydroxyl group or the carboxyl group partially remains as such.

[0046] The ester of aliphatic monocarboxylic acids (fatty acids) (III-1) is an ester of one or more selected from the above-mentioned aliphatic monocarboxylic acids(fatty acids) (II-1) and one or more selected from the above-mentioned mono, di, tri or higher polyalcohols (I-1 to I-3) . Examples of the esters are aliphatic monocarboxylic acids. Concrete examples of the esters are glycerin monooleate, glycerin dioleate, sorbitan monooleate, sorbitan dioleate, and the like.

[0047] As esters (3-1) other than such aliphatic acid ester base ashless friction modifier, aliphatic esters having straight-chain or branched hydrocarbons of C 1 to 5 of C31 to 40 are exemplified, and esters composed of aliphatic acids having such a hydrocarbon and aliphatic monoalcohols or aliphatic polyalcohols are exemplified. Among them, the esters having kinetic viscosity of 1 to 100mm² at 100°C can be used as lubricating oil base oil, and normally can be distinguished from the aliphatic acid ester group ashless friction modifier. As these examples, tri or more polyols such as trimethyl propane caprylate, trimethyl propane pelargonate, pentaerythritol 2-ethylhexanoate, and pentaerythritol pelargonate of C3 to 40 preferably C4 to 18, more preferably 4 to 12, tri or more polyols having a neopentyl structure, one kind, two kinds or more of single esters selected of monocarboxylate acid of C1 to 40, preferably C 4 to 18, more preferably C6 to 12, polyol esters such as complex esters and the mixtures, or esters added by alkylene oxide are exemplified. They may be complete esters by esterifying all of hydroxyl groups or carboxyl groups or partial esters where hydroxyl groups or the carboxyl groups partially remain, but the complete esters are preferable and the hydroxyl bases are normally equal to or less than 100mgKOH/g, preferably 50mgKOH/g, more preferably 10mgKOH/g. Among them, the esters having kinetic viscosity of preferably 2 to 60mm² , more preferably 3 to 50mm²at

100°C can be used as lubricating oil base oil.

[0048] The ester of aliphatic polycarboxylic acids (III-2) is an ester of one or more selected from the above-mentioned aliphatic polycarboxylic acids (II-1) and one or more selected from the above-^L mentioned mono, di, tri or higher polyalcohols (I-1 to I-3). Its preferred examples are diesters of one or more polycarboxylic acid selected from dicarboxylic acids having from 2 to 40, preferably from 4 to 18, more preferably from 6 to 12 carbon atoms, and one or more selected from monoalcohols having from 4 to 40, preferably from 4 to 18, more preferably from 6 to 14, such as dibutyl maleate, ditridecyl glutamate, di-2-ethylhexyl adipate, diisodecyladipate, ditridecyladipate, di-2-ethylhexylsebacate, and copolymers of these diesters (e.g., dibutyl maleate) and poly- α -olefins having from 4 to 16 carbon atoms; and esters of α -olefin adducts to acetic anhydride or the like, and alcohols having from 1 to 40 carbon atoms. Among them, the esters having kinetic viscosity of 1 to 100mm² at 100°C can be used as lubricating oil base oil.

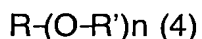
[0049] The ester of carbon-cyclic carboxylic acids (III-3) is an ester of one or more selected from the above-mentioned carbon-cyclic carboxylic acids (II-3), and one or more selected from the above-^L mentioned mono, di, tri or higher polyalcohols (I-1 to I-3). Its preferred examples are aromatic carboxylic acid ester such as phthalic acid ester, trimellitic acid ester, pyromellitic acid ester, salicylic acid ester. Among them, the esters having kinetic viscosity of 1 to 100mm² at 100°C can be used as lubricating oil base oil.

[0050] The ester of heterocyclic carboxylic acids (III-4) is an ester of one or more selected from the above-mentioned heterocyclic carboxylic acids (II-4), and one or more selected from the above-mentioned mono, di, tri or higher polyalcohols (I-1 to I-3). Among them, the esters having kinetic viscosity of 1 to 100mm² at 100°C can be used as lubricating oil base oil.

[0051] As alkylene oxide additives (III-5) of alcohols or esters, esters by adding alkylene oxides to one or more kinds selected of the above mono or more polyalcohols (I-1 to I-3) or esters by adding the alkylene oxides to the above (III-1 to 4) esters are exemplified. Among them, the esters having kinetic viscosity of 1 to 100mm² at 100°C can be used as lubricating oil base oil.

[0052] The above-mentioned ethers (IV) are compounds represented by the following general formula (4).

[0053]



[0054]

for example, a compound containing one or more ether bonds.

[0055] Examples of ethers (IV) are mentioned below:

Saturated or unsaturated aliphatic ethers (IV-1);

Aromatic ethers (IV-2);

Cyclic ethers (IV-3);

Polyalcoholic esters (IV-4); and

Mixtures of two or more selected from the above three kinds of ethers (IV-5) .

[0056] Saturated or unsaturated aliphatic ethers(aliphatic monoethers) are concretely saturated or unsaturated aliphatic ethers (IV-1) having from 1 to 40 carbon atoms (in which the saturated or unsaturated aliphatic structure may be linear or branched, and the unsaturated bond may be in any desired position) such as dimethyl ether, diethyl ether, di-n-propyl ether, diisopropyl ether, dibutyl ether, diisobutyl ether, di-n-amyl ether, diisoamyl ether, dihexyl ether, diheptyl ether, dioctyl ether, dinonyl ether, didecyl ether, diundecyl ether, didodecyl ether, ditridecyl ether, ditetradecyl ether, dipentadecyl ether, dihexadecyl ether, diheptadecyl ether, dioctadecyl ether, dinonadecyl ether, dieicosyl ether, methyl ethyl ether, methyl n-propyl ether, methyl isopropyl ether, methyl isobutyl ether, methyl tert-butylether, methyl n-amyl ether, methyl isoamyl ether, ethyl n-propyl ether, ethyl isopropyl ether, ethyl isobutyl ether, ethyl tert-butyl ether, ethyl n-amyl ether, ethyl isoamyl ether, divinyl ether, diallyl ether, methyl vinyl ether, methyl allyl ether, ethyl vinyl ether, ethyl allyl ether.

[0057] Concretely, aromatic ethers (IV-2) include, for example, anisole, phenetole, phenyl ether, benzyl ether, phenyl benzyl ether, a-naphthyl ether, b-naphthyl ether, polyphenyl ether, perfluoroether; and these may have a saturated or unsaturated group (in which the saturated or unsaturated group may be linear or branched, and the unsaturated bond may be in any desired position, and the number and the position of the substituents are not defined) . Preferably, these are liquid under the service condition thereof, especially at room temperature.

[0058] Concretely, cyclic ethers (IV-^L 3) are those having from 2 to 40 carbon atoms, including, for example, ethylene oxide, propylene oxide, trimethylene oxide, tetrahydrofuran, tetrahydropyran,

dioxane, glycidyl ether; and these may have a saturated or unsaturated group, a carbon ring, or a saturated or unsaturated aliphatic group^L having carbon ring (in which the saturated or unsaturated group may be linear or branched, and the unsaturated bond may be in any desired position, and the number and the position of the substituents are not defined).

Ethers of polyalcohols (IV^L 4) include one or not less than two ethers of one or not less than two polyalcohols selected from the above^L mentioned dialcohols or trialcohols (I^L 2 to I^L 2) or Monoalcohols . The ethers referred to herein may be complete ethers in which all hydroxyl groups of polyalcohols are etherified, or partial ethers in which a part of hydroxyl groups remains, in which the partial ethers are preferable since they exhibit a low friction properties.

[0059] The ketones (V) are compounds represented by the following general formula (5):

[0060]



[0061]

, for example, an oxygen-containing organic compound and a compound containing one or more carbonyl bonds.

[0062] Specific examples of the aforementioned ketones (V) include: a saturated or unsaturated aliphatic ketone (V-1);

a carbon-ring ketone (V-2);

a heterocyclic ketone (V-3);

a ketone alcohol (V-4);

a ketone acid (V-5); and

a mixture of two or more selected from ketones of the aforementioned five kinds of ketones (V-6).

[0063] The saturated or unsaturated aliphatic ketone (V-1) can specifically be a saturated or unsaturated aliphatic ketone with 1 to 40 carbon atoms (which may be linear or ramified or branched and may have an unsaturated bond in an arbitrary position), such as acetone, methyl ethyl ketone, methyl propyl ketone, methyl isopropyl ketone, methyl butyl ketone, methyl isobutyl ketone, pinacolone, diethyl ketone, butyrone, diisopropylketone, methyl vinyl ketone, mesityl oxide or methyl heptenone.

[0064] Also the carbon-ring ketone (V-2) can specifically be a carbon-^L ring ketone with 1 to 40 carbon atoms such as cyclobutanone, cyclopentanone, cyclohexanone, acetophenone, propiophenone, butyrophenone, valerophenone, benzophenone,

dibenzylketone or 2-acetonaphthone, which may have a saturated or unsaturated aliphatic group (that may be linear or ramified, arbitrary in the position of an unsaturated bond and also arbitrary in the position and number of substitution).

[0065] Also the heterocyclic ketone (V-3) can specifically be a carbocyclic ketone with 1 to 40 carbon atoms such as acetothienone or 2-acetofuron, which may have a saturated or unsaturated aliphatic group (that may be linear or ramified, arbitrary in the position of an unsaturated bond and also arbitrary in the position and number of substitution).

[0066] Also the ketone alcohol (ketol) (V-4) can specifically be a ketone alcohol with 1 to 40 carbon atoms such as acetol, acetoin, acetoethyl alcohol, diacetone alcohol, phenacyl alcohol or benzoin, which may have a carbon ring or a hetero ring (heterocycle) or may have a carbon ring or a hetero ring having a saturated or unsaturated aliphatic group (that may be linear or ramified, arbitrary in the position of an unsaturated bond and also arbitrary in the position and number of substitution).

[0067] Also the ketone acid (V-5) can specifically be a ketone acid with 1 to 40 carbon atoms for example an α -ketone acid such as piruvic acid, benzoylformic acid, or phenylpiruvic acid, a β -ketone acid such as acetoacetic acid, propionylacetic acid or benzoylacetic acid, or a γ -ketone acid such as levulinic acid or β -benzoylpropionic acid.

[0068] The aldehydes (VI) are oxygen-containing organic compounds represented by the following general formula (6):

[0069]



[0070], for example, a compound having one or two aldehyde groups.

[0071] Specific examples of the aforementioned aldehydes include (VI):

a saturated or unsaturated aliphatic aldehyde (V1-1);

a carbon-ring aldehyde (V1-2)

a heterocyclic aldehyde (V1-3); and

a mixture of two or more selected from the aldehydes of the aforementioned three kinds of aldehydes (V1-4).

[0072] The saturated or unsaturated aliphatic aldehyde (V1-1) can specifically be a saturated or unsaturated aliphatic aldehyde with 1 to 40 carbon atoms (such saturated or unsaturated aliphatic structure may be linear or ramified with an arbitrary position in an unsaturated bond) such as formaldehyde, acetaldehyde, propionaldehyde, butyl aldehyde, isobutyl aldehyde, valeric aldehyde, isovaleraldehyde, pivalicaldehyde, caproicaldehyde, heptonic aldehyde, capryl aldehyde, peralgonic aldehyde, capric aldehyde, undecyl aldehyde, lauric aldehyde, tridecyl aldehyde, myristic aldehyde, pentadecyl aldehyde, palmitic aldehyde, margaricaldehyde, stearicaldehyde, acrolein, crotonaldehyde, propionic aldehyde, glyoxal or succinic aldehyde.

[0073] The carbon-ring aldehyde (V1-2) can specifically be a carbon-ring aldehyde with 1 to 40 carbon atoms such as benzaldehyde, o-toluic aldehyde, m-toluic aldehyde, p-toluic aldehyde, salicyl aldehyde, cinnamic aldehyde, a-naphthoic aldehyde, or b-naphthoic aldehyde, which may have a saturated or unsaturated aliphatic group (that may be linear or ramified, arbitrary in the position of an unsaturated bond and also arbitrary in the position and number of substitution).

[0074] The heterocyclic aldehyde (V1-3) can specifically be a heterocyclic aldehyde with 1 to 40 carbon atoms such as furfural, which may have a saturated or unsaturated aliphatic group (that may be linear or ramified, arbitrary in the position of an unsaturated bond and also arbitrary in the position and number of substitution).

[0075] The carbonates (V11) are oxygen-^Lcontaining organic compounds represented by the following general formula (7):

[0076]



[0077] for example, a compound having one or two carbonate bonds.

[0078] [0079]The carbonates (VII) can specifically be a carbonate with 1 to 40 carbon atoms having a saturated or unsaturated aliphatic group, a carbon-ring group, a carbon-ring group having a saturated or unsaturated aliphatic group, or a saturated or unsaturated aliphatic group having a carbon-ring group (such saturated or unsaturated aliphatic group being linear or ramified, arbitrary in the position of an unsaturated bond and also arbitrary in the position and number of substitution), such as dimethyl carbonate, diethylcarbonate,

di-n-propylcarbonate, diisopropylcarbonate, di-n-butyl carbonate, diisobutyl carbonate, di-tert-butyl carbonate, dipentyl carbonate, dihexyl carbonate, diheptyl carbonate, dioctyl carbonate, dinonyl carbonate, didecyl carbonate, diundecyl carbonate, didodecyl carbonate, ditridecyl carbonate, ditetradecyl carbonate, dipentadecyl carbonate, dihexadecyl carbonate, diheptadecyl carbonate, dioctadecyl carbonate, or diphenyl carbonate, or a hydroxy (poly) oxyalkylene carbonate formed by adding an alkylene oxide to such carbonate.

[0080] On the other hand, R and R' in the general formula (1) - (7) each independently represents a hydrocarbon group such as an alkyl group, an alkenyl group, an alkylene group, a cycloalkyl group, an alkylcycloalkyl group, an aryl group, an alkylaryl group, or an arylalkyl group (such hydrocarbon group may further contain one or more kinds of group or bond selected from a hydroxyl group, a carboxyl group, a carbonyl group, an ester bond and an ether bond, and may further contain an element other than carbon, hydrogen and oxygen, such as nitrogen or sulfur (for example a heterocyclic compound), a halogen (such as fluorine or chlorine), phosphorus, boron or a metal. The hydrocarbon group is not particularly restricted in the number of carbon atoms, but preferably has 1 to 40 carbon atoms, more preferably 2 to 30 carbon atoms and particularly preferably 3 to 20 carbon atoms.

[0081] Examples of the alkyl group include an alkyl group with 1 to 40 carbon atoms such as a methyl group, an ethyl group, an n-propyl group, an isopropyl group, an n-butyl group, an isobutyl group, a sec-butyl group, a tert-butyl group, a linear or ramified pentyl group, a linear or ramified hexyl group, a linear or ramified heptyl group, a linear or ramified octyl group, a linear or ramified nonyl group, a linear or ramified decyl group, a linear or ramified undecyl group, a linear or ramified dodecyl group, a linear or ramified tridecyl group, a linear or ramified tetradecyl group, a linear or ramified pentadecyl group, a linear or ramified hexadecyl group, a linear or ramified heptadecyl group, a linear or ramified octadecyl group, a linear or ramified nonadecyl group, a linear or ramified icosyl group, a linear or ramified hencosyl group, a linear or ramified docosyl group, a linear or ramified tricosyl group, or a linear or ramified tetracosyl group, preferably an alkyl group with 2 to 30 carbon atoms and particularly preferably an alkyl group with 3 to 20 carbon atoms.

[0082] Examples of the alkenyl group include an alkenyl group with 2 to 40 carbon atoms such as a vinyl group, a linear or ramified propenyl group, a linear or ramified butenyl group, a linear or ramified pentenyl group, a linear or

ramified hexenyl group, a linear or ramified heptenyl group, a linear or ramified octenyl group, a linear or ramified nonenyl group, a linear or ramified decenyl group, a linear or ramified undecenyl group, a linear or ramified dodecenyl group, a linear or ramified tridecenyl group, a linear or ramified tetradecenyl group, a linear or ramified pentadecenyl group, a linear or ramified hexadecenyl group, a linear or ramified heptadecenyl group, a linear or ramified octadecenyl group, a linear or ramified nonadecenyl group, a linear or ramified icosenyl group, a linear or ramified heneicosenyl group, a linear or ramified docosenyl group, a linear or ramified tricosenyl group, or a linear or ramified tetracosenyl group, preferably an alkenyl group with 2 to 30 carbon atoms and particularly preferably an alkenyl group with 3 to 20 carbon atoms.

[0083] Examples of the cycloalkyl group include a cycloalkyl group with 3 to 40 carbon atoms such as a cyclopentyl group, a cyclohexyl group, a cycloheptyl group or a cyclooctyl group, preferably a cycloalkyl group with 3 to 20 carbon atoms and particularly preferably a cycloalkyl group with 5 to 8 carbon atoms.

Examples of the alkylcycloalkyl group include an alkylcycloalkyl group with 4 to 40 carbon atoms such as a methylcyclopentyl group, a dimethylcyclopentyl group (including all structural isomers), a methylethylcyclopentyl group (including all structural isomers), a diethylcyclopentyl group (including all structural isomers), a methyl cyclohexyl group, a dimethylcyclohexyl group (including all structural isomers), a methylethylcyclohexyl group (including all structural isomers), a diethylcyclohexyl group (including all structural isomers), a methylcycloheptyl group, a dimethylcycloheptyl group (including all structural isomers), a methylethylcycloheptyl group (including all structural isomers), or a diethylcycloheptyl group (including all structural isomers), preferably an alkylcycloalkyl group with 5 to 20 carbon atoms, particularly preferably an alkylcycloalkyl group with 6 to 12 carbon atoms.

[0084] Examples of the aryl group include an aryl group with 6 to 20 carbon atoms such as a phenyl group or a naphthyl group, preferably an aryl group with 6 to 10 carbon atoms.

Examples of the alkylaryl group include an alkylaryl group with 7 to 40 carbon atoms for example a monosubstituted phenyl group such as a tolyl group (including all structural isomers), an ethylphenyl group (including all structural isomers), a linear or ramified propylphenyl group (including all structural isomers), a linear or ramified butylphenyl group (including all structural isomers), a linear or ramified pentylphenyl group (including all structural isomers), a linear or

ramified hexylphenyl group (including all structural isomers), a linear or ramified heptylphenyl group (including all structural isomers), a linear or ramified octylphenyl group (including all structural isomers), a linear or ramified nonylphenyl group (including all structural isomers), a linear or ramified decylphenyl group (including all structural isomers), a linear or ramified undecylphenyl group (including all structural isomers), or a linear or ramified dodecylphenyl group (including all structural isomers); or an aryl group having two more, same or different linear or ramified alkyl groups such as a xylyl group (including all structural isomers), a diethylphenyl group, a dipropylphenyl group, a 2-methyl-6-tert-butylphenyl group, a 2,6-di-tert-butyl-4-methylphenyl group, or a 2,6-di-tert-butyl-4-(3,5-di-tert-butyl-4-benzyl) phenyl group (alkyl group may further include an aryl group, an alkylaryl group or an arylalkyl group including all structural isomers), preferably an alkylaryl group with 7 to 20 carbon atoms and particularly preferably an alkylaryl group with 7 to 12 carbon atoms.

[0085] Also examples of the arylalkyl group include an arylalkyl group with 7 to 40 carbon atoms such as a benzyl group, a phenylethyl group, a phenylpropyl group (including isomers of propyl group), a phenylbutyl group (including isomers of butyl group), a phenylpentyl group (including isomers of pentyl group) or a phenylhexyl group (including isomers of hexyl group), preferably an arylalkyl group with 7 to 20 carbon atoms and particularly preferably an arylalkyl group with 7 to 12 carbon atoms.

[0086] Derivatives of the above-mentioned organic oxygen-containing organic compounds (C) can be used like the oxygen-containing organic compounds. Examples of the derivatives are nitrogen-containing compounds, sulfur or sulfur containing compound, boron-containing compound, halogen elements or halogen element-containing compounds, metal elements or metal-containing compounds (organic or inorganic ones), and compounds obtained by reacting alkylene oxide; however, the derivatives are not limited to the above ones. The derivatives concretely include, for example, compounds prepared by sulfidizing one selected from the above-mentioned alcohols, carboxylic acids, esters and ethers, ketones, aldehydes and carbonates; compounds prepared by halogenating (fluorinating, chlorinating) the same one; its reaction products with acids, such as sulfuric acid, nitric acid, boric acid, phosphoric acid, or their esters or metal salts; and its reaction products with metals, metal-containing compounds or amine compounds. Of those, preferred are reaction products of one or more selected from alcohols and carboxylic acids and their derivatives,

with amine compounds (e.g., Mannich reaction products, acylated products, amides).

[0087] The amine compounds as referred to herein include ammonia, monoamines, diamines, and polyamines. More concretely, their examples are ammonia; alkylamines having an alkyl group with from 1 to 30 carbon atoms (in which the alkyl group may be linear or branched) such as methylamine, ethylamine, propylamine, butylamine, pentylamine, hexylamine, heptylamine, octylamine, nonylamine, decylamine, undecylamine, dodecylamine, tridecylamine, tetradecylamine, pentadecylamine, hexadecylamine, heptadecylamine, octadecylamine, stearylamine, dimethylamine, diethylamine, dipropylamine, dibutylamine, dipentylamine, dihexylamine, diheptylamine, dioctylamine, dinonylamine, didecylamine, diundecylamine, didodecylamine, ditridecylamine, ditetradecylamine, dipentadecylamine, dihexadecylamine, diheptadecylamine, dioctadecylamine, methylethylamine, methylpropylamine, methylbutylamine, ethylpropylamine, ethylbutylamine, and propylbutylamine; alkenylamines having an alkenyl group with from 2 to 30 carbon atoms (in which the alkenyl group may be linear or branched) such as ethenylamine, propenylamine, butenylamine, octenylamine, and oleylamine; alkanolamines having an alkanol group with from 1 to 30 carbon atoms (in which the alkanol group may be linear or branched) such as methanolamine, ethanolamine, propanolamine, butanolamine, pentanolamine, hexanolamine, heptanolamine, octanolamine, nonanolamine, methanolethanolamine, methanolpropanolamine, methanolbutanolamine, ethanolpropanolamine, ethanolbutanolamine, and propanolbutanolamine; alkylenediamines having an alkylene group with from 1 to 30 carbon atoms, such as methylenediamine, ethylenediamine, propylenediamine, and butylenediamine; polyamines such as diethylenetriamine, triethylenetetramine, tetraethylenepentamine, pentaethylenehexamine; compounds derived from the above-mentioned monoamines, diamines or polyamines and further having an alkyl or alkenyl group with from 8 to 20 carbon atoms, such as undecyldiethylamine, undecyldiethanolamine, dodecyldipropanolamine, oleyldiethanolamine, oleylpropylenediamine, stearyl tetraethylenepentamine; heterocyclic compounds such as N-hydroxyethyloleylimidazoline; alkylene oxide

adducts of these compounds; and their mixtures. Of those nitrogen-^L containing compounds, preferred are aliphatic amines having an alkyl or alkenyl group with from 10 to 20 carbon atoms (these may be linear or branched) such as decylamine, dodecylamine, tridecylamine, heptadecylamine, octadecylamine, oleylamine and stearylamine. Of the derivatives of these oxygen-containing organic compounds, preferred are amides of carboxylic acid having 8 to 20 carbon atoms, of the above-mentioned aliphatic monocarboxylic acids (fatty acids) (II-1) such as oleic amide.

[0088] The contained amount of the oxygen-containing organic compound (C) or the aliphatic amine compound (D) is not particularly restricted; however, the lower limit of the contained amount based on the total amount (total mass) of the low-friction agent composition is normally 0.001 %, preferably 0.05 %, morepreferably 0.1%, particularly preferably 0.5% from the viewpoint of friction reduction effect. The upper limit of the contained amount is 100 % as mentioned above. In case that other components, particularly the medium is blended, the upper limit based on the total amount of the low-^L friction agent composition is normally 50 %, preferably 20 %m, more preferably 10 %, particularly preferably 5 % from the viewpoint of solubility to the medium and storage stability. In the present invention, even addition of the oxygen-^L containing organic compound (C) and/or the aliphatic amine compound (D) in a small amount of about 0.1 to 2 % can exhibit an excellent low friction characteristics.

Additionally, if the solubility and storing stability of the aliphatic amine compound (D) to the medium is taken into serious consideration, it is preferable that the upper limit is preferably 3.0 %, more preferably 2.0 %, further preferably 1.4 %.

[0089]As the above-^L mentioned medium, concrete examples are mineral oil, synthetic oil, natural fat and oil, diluted oil, grease, wax, hydrocarbons having the carbon number of 3 to 40, hydrocarbon solvents, organic solvents other than the hydrocarbon solvents, water and the like, and a mixture of these, and particularly these in the state of liquid, grease or wax under a sliding condition and at a normal temperature. As the above-^L mentioned medium there is particularly preferably employed a lubricating oil base oil (a base oil of the lubricating oil). Such lubricating oil base oil is not particularly limited and any ordinary base oil, either mineral oil type or synthetic type, for lubricant composition can be employed.

[0090] Examples of the lubricating oil base oil of mineral oil type include a product formed by subjecting a lubricant fraction, obtained as a result of

atmospheric distillation or vacuum distillation of crude oil, to at least one of solvent deasphalting, solvent extraction, hydrocracking, solvent dewaxing, hydrorefining and wax isomerization, particularly a base oil subjected to hydrocracking, hydrorefining or wax isomerization. Among such products, there is preferred a mineral oil obtained by hydrorefining or hydrocracking, or an isoparaffinic mineral oil obtained by isomerization of GTL (gas-to-liquid) wax by a Fischer-Tropsche process or of normal paraffin-rich wax obtained in a dewaxing step of lubricant oil.

[0091] Examples of the lubricating oil base oil of synthetic type include an alkylnaphthalene, an alkylbenzene, a polybutene and a hydrogenated product thereof; a poly- α -olefin such as a 1-octene oligomer, a 1-decene oligomer, an ethylene propylene oligomer or a hydrogenated product thereof; an isobutene oligomer and a hydrogenated product of isobutene oligomer; isoparaffin, alkylbenzene, alkylnaphthalene, diester (for example, trimethylpropane ester such as trimethylpropane caprylate, trimethylpropane pelargonate, trimethylolpropane pelargonate, trimethylolpropane isostearinate and the like; and pentaerythritol ester such as pentaerythritol- ω -2-ethylhexanoate, pentaerythritol pelargonate), polyoxyalkylene glycol, dialkyldiphenyl ether, polyphenyl ether and the like; and mixtures of these. Preferable examples of the synthetic lubricating oil base oil are poly- α -olefin such as 1-octene oligomer, 1-deceneoligomer and the like, and hydrogenated product thereof.

[0092] In addition to the use of the lubricating oil base oil of mineral oil type or the lubricating oil base oil of synthetic type either singly or as a mixture, it is also possible to use a mixture of two or more kinds of the base oil of mineral oil type or the base oil of synthetic type. Also in such mixture, a mixing ratio of two or more kinds of the base oils is not particularly restricted and can be selected arbitrarily.

[0093] A total aromatic content of the lubricating oil base oil is not particularly restricted, but is preferably 15 % or less, more preferably 10 % or less and further preferably 8 %. A total aromatic content in the lubricating oil base oil exceeding 15 % results in an inferior stability to oxidation and is undesirable. A composition of a high friction reducing effect can be obtained even with a total aromatic content in the lubricating oil base oil of 2 % or less, or even 0 %, for example of lubricating oil base oil such as a highly hydrocracking mineral oil, a wax isomerized mineral oil, a poly- α -olefin or a hydrogenated product,

a hydrogenated product of 1-decene oligomer, polyole ester, or a mixture thereof. In case the content of the organic oxygen-containing organic compound (C) (excluding esters as the lubricating oil base oil) is higher (for example, at 2 % or higher, since the storage stability may be deteriorated, it is preferable, if necessary, to regulate the total aromatic content (for example at 2% or higher) of the lubricating oil base oil by blending a solvent extracted mineral oil or an alkylbenzene, or to employ an ester as the lubricating oil base oil, thereby raising the solubility of the oxygen containing compound (C). The "total aromatic content" means a content of an aromatic fraction measured according to ASTM D2549, and such aromatic fraction ordinarily contains alkylbenzenes, alkylnaphthalenes, anthracene, phenanthrene, alkylated substances thereof, a compound in which four or more benzene rings are condensed, and compounds containing heteroaromatic structure such as pyridines, quinolines, phenols and naphthols.

[0094] Also the lubricating oil base oil is not particularly restricted in a kinetic viscosity thereof, but in case of use as a lubricant composition for an internal combustion engine, a kinetic viscosity at 100°C is preferably 2 mm²/s or higher, and more preferably 3 mm²/s or higher. Also an upper limit is preferably 20 mm²/s or less, more preferably 10 mm²/s or less and particularly preferably 8 mm²/s or less. A lubricating oil base oil with a kinetic viscosity at 100°C of 2 mm²/s or higher allows to obtain a composition capable of sufficient oil film formation, an excellent lubricating property and a smaller evaporation loss of the base oil under a high temperature condition. On the other hand, a kinetic viscosity at 100°C of 20 mm²/s or less reduces a fluid resistance, thereby allowing to obtain a composition with a smaller frictional resistance in a lubricated site. If the kinetic viscosity is less than 2 mm²/s, there is the possibility that a sufficient frictional resistance can be obtained while a vaporizing characteristics is inferior, which is not preferable. If the kinetic viscosity exceeds 20 mm²/s, there is the possibility that a low friction characteristics is difficult to be exhibited while a low temperature characteristics are degraded, which are not preferable. In the present invention, a mixture which are prepared by freely mixing two or more base oils selected from the above-mentioned base oils can be used, in which the base oils having the kinetic viscosity (as a single base oil) other than the above-mentioned can be also used as far as the base oils have a kinetic viscosity at 100°C which viscosity is within the above-mentioned preferable range.

[0095] Furthermore, a viscosity index of the lubricating oil base oil is not particularly restricted but is preferably 80 or higher, and, in case of use as a lubricant composition for an internal combustion engine, it is preferably 100 or higher, more preferably 120 or higher, and maybe within a range of 140 to 250. A lubricating oil base oil of a high viscosity index allows to obtain a composition excellent not only in a low-temperature viscosity characteristics but also in a less oil consumption, a fuel efficiency characteristics, and a friction reducing effect.

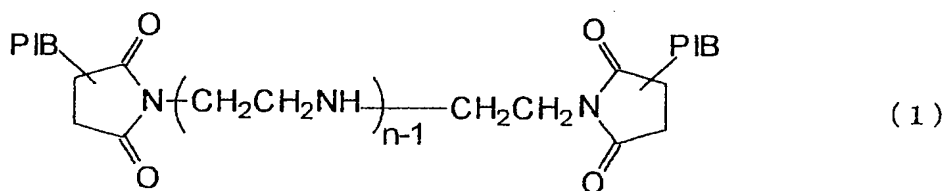
[0096] The low-friction agent composition may further include an ashless dispersant, an abrasion preventing agent or an extreme pressure agent, a metal-^L based detergent, an antioxidant, a viscosity index improver, a friction modifier agent other than (C) and (D), an antirusting agent, a nonionic surfactant, an antiemulsifier agent, a metal deactivator, or a defoaming agent singly or in a combination of plural kinds, for improving required performances.

[0097] As the ashless dispersant, various known ashless dispersants can be employed. For example polybutenylsuccinimide or a derivative thereof can be advantageously employed.

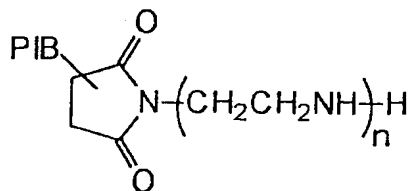
The above-^L mentioned polybutenyl succinimide include compounds represented by the following chemical formulas

(1) and (2).

[0098]



[0099]



(2)

[0100] PIB in these chemical formulae represents a polybutenyl group derived from polybutene. The polybutene can be prepared by polymerizing high-purity isobutene or a mixture of 1-butene and isobutene in the presence of a boron fluoride catalyst or an aluminum chloride catalyst in such a manner that the polybutene attains a number-average molecular weight of 900 to 3,500, preferably 1,000 to 2,000. When the number-average molecular weight of the polybutene is less than 900, there is a possibility of failing to attain a sufficient detergent effect. When the number-average molecular weight of the polybutene exceeds 3,500, the polybutene may undesirably deteriorate in low-temperature fluidity.

In each of the chemical formulae, n represents an integer of 1 to 5, preferably 2 to 4, so as to attain a good detergent effect. In the production of the polybutenyl succinimide, the polybutene may be used after purified by removing trace amounts of fluorine and chlorine residues, which result from the above polybutene production catalyst, by any suitable treatment (such as adsorption process or washing process). The amount of the fluorine and chlorine residues is preferably controlled to 50 ppm or less, more preferably 10 ppm or less, most preferably 1 ppm or less.

[0101] The production method of the polybutenyl succinimide is not particularly restricted. For example, the polybutenyl succinimide can be prepared by reacting an chloride of the above-mentioned polybutene, or the polybutene from which fluorine and chlorine residues are removed, with maleic anhydride at 100 to 200°C to form polybutenyl succinate, and then, reacting the thus-formed polybutenyl succinate with polyamine (such as diethylene triamine, triethylene tetramine, tetraethylene pentamine or pentaethylene hexamine).

[0102] The polybutenyl succinimide derivative can be exemplified by boron- and acid-modified compounds obtained by reacting the polybutenyl succinimide of

the formulas (1) and (2) with boron compounds or oxygen-containing organic compounds so as to neutralize or amidate the whole or part of the remaining amino and/or imide groups. Among these, boron-containing polybutenyl succinimide, especially boron-containing bis(polybutenyl) succinimide, is preferably used.

[0103] The above boron compound can be a boric acid, a borate or a boric acid ester. Specific examples of the boric acid include orthoboric acid, metaboric acid and paraboric acid. Specific examples of the borate include: ammonium salts including ammonium borates, such as ammonium metaborate, ammonium tetraborate, ammonium pentaborate and ammonium octaborate. Specific examples of the boric acid ester include: esters of boric acids and alkylalcohols (preferably C1-C6 alkylalcohols), such as monomethyl borate, dimethyl borate, trimethyl borate, monoethyl borate, diethyl borate, triethyl borate, monopropyl borate,^L dipropyl borate, tripropyl borate, monobutyl borate, dibutyl borate and tributyl borate. Herein, the content ratio of nitrogen to boron (B/N) by mass in the boron-containing polybutenyl succinimide is usually 0.1 to 3, preferably 0.2 to 1.

The above oxygen-containing organic compound can be exemplified by: C1-C30 monocarboxylic acids, such as formic acid, acetic acid, glycolic acid, propionic acid, lactic acid, butyric acid, valeric acid, caproic acid, enanthic acid, caprylic acid, pelargonic acid, capric acid, undecylic acid, lauric acid, tridecanoic acid, myristic acid, pentadecanoic acid, palmitic acid, margaric acid, stearic acid, oleic acid, nonadecanoic acid and eicosanoic acid; C2-C30 polycarboxylic acids, such as oxalic acid, phthalic acid, trimellitic acid and pyromellitic acid, and anhydrides and esters thereof; C2-C6 alkylene oxides; and hydroxyl (poly) oxyalkylene carbonates.

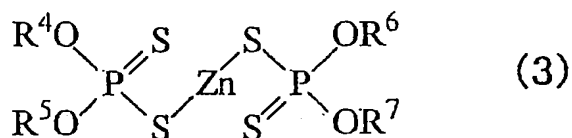
[0104] The amount of the polybutenyl succinimide and/or the derivative thereof added in the low-friction agent composition is not particularly restricted, and is preferably 0.1 to 15%, more preferably 1.0 to 12%, based on the total amount of the lubricating oil. When the amount of the polybutenyl succineimide and/or the derivative thereof is less than 0.1%, there arises a possibility of failing to attain a sufficient detergent effect. It becomes uneconomical when the amount of the polybutenyl succineimide and/or the derivative thereof exceeds 15%. In addition, such a large amount of the polybutenyl succineimide and/or the derivative thereof tends to cause a deterioration in demulsification ability.

The ashless dispersant other than the above-mentioned can be exemplified by polybutenylbenzylamines and polybutenylamines each having polybutenyl

groups of number-average molecular weight of 900 to 3,500, polybutenyl succinimides having polybutenyl groups of number-average molecular weight of less than 900 and derivatives thereof.

[0105] As an antioxidant and as an anti-wear agent, various known ones can be blended.

[0106] For example, it is preferable to contain zinc dithiophosphate represented by the following chemical formula (3) as an antioxidant and as an anti-wear agent.



[0107] In the chemical formula (3), R⁴, R⁵, R⁶ and R⁷ each represent C1-C24 hydrocarbon groups. The C1-C24 hydrocarbon group is preferably a C1-C24 straight-chain or branched-chain alkyl group, a C3-C24 straight-chain or branched chain alkenyl group, a C5-C13 cycloalkyl or straight-chain or branched-chain alkylcycloalkyl group, a C6-C18 aryl or straight-chain or branched-chain alkylaryl group, or a C7-C19 arylalkyl group. The above alkyl group or alkenyl group can be primary, secondary or tertiary.

[0108] Specific examples of R⁴, R⁵, R⁶ and R⁷ include: alkyl groups, such as methyl, ethyl, propyl, butyl, pentyl, hexyl, heptyl, octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, nonadecyl, icosyl, heneicosyl, docosyl, tricosyl and tetracosyl; alkenyl groups, such as propenyl, isopropenyl, butenyl, butadienyl, pentenyl, hexenyl, heptenyl, octenyl, nonenyl, decenyl, undecenyl, dodecenyl, tridecenyl, tetradecenyl, pentadecenyl, hexadecenyl, heptadecenyl, octadecenyl (oleyl), nonadecenyl, icosenyl, heneicosenyl, docosenyl, tricosenyl and tetracosenyl; cycloalkyl groups, such as cyclopentyl, cyclohexyl and cycloheptyl; alkylcycloalkyl groups, such as methylcyclopentyl,

dimethylcyclopentyl, ethylcyclopentyl, propylcyclopentyl, ethylmethylcyclopentyl, trimethylcyclopentyl, diethylcyclopentyl, ^L ethyldimethylcyclopentyl, propylmethylcyclopentyl, propylethylcyclopentyl, di-^L propylcyclopentyl, propyl ethylmethylcyclopentyl, methylcyclohexyl, dimethylcyclohexyl, ethylcyclohexyl, propylcyclohexyl, ethylmethylcyclohexyl, trimethylcyclohexyl, diethylcyclohexyl, ethyldimethylcyclohexyl, propylmethylcyclohexyl, propylethylcyclohexyl, di-^L propylcyclohexyl, propylethylmethylcyclohexyl, methylcycloheptyl, dimethylcycloheptyl, ethylcycloheptyl, propylcycloheptyl, ethylmethylcycloheptyl, trimethylcycloheptyl, diethylcycloheptyl, ethyldimethylcycloheptyl, propylmethylcycloheptyl, propylethylcycloheptyl, di-^L propylcycloheptyl and propylethylmethylcycloheptyl; aryl groups, such as phenyl and naphthyl; alkylaryl groups, such as tolyl, xylyl, ethylphenyl, propylphenyl, ethylmethylphenyl, trimethylphenyl, butylphenyl, propylmethylphenyl, diethylphenyl, ethyldimethylphenyl, tetramethylphenyl, pentylphenyl, hexylphenyl, heptylphenyl, octylphenyl, nonylphenyl, decylphenyl, undecylphenyl and dodecylphenyl; and arylalkyl groups, such as benzyl, methylbenzyl, dimethylbenzyl, phenethyl, methylphenethyl and dimethylphenethyl.

The above-^L mentioned hydrocarbon groups include all considerable straight or branched chain structures. The position of double bond of alkenyl group, the bonding position of alkyl group to cycloalkyl group and the bonding position of alkyl group to aryl group are free.

[0109] Specific examples of the zinc dithiophosphate usable include zinc diisopropyldithiophosphate, zinc diisobutyldithiophosphate, zinc di-sec-butyldithiophosphate, zinc di-sec-pentyldithiophosphate, zinc di-n-hexyldithiophosphate, zinc di-sec-hexyldithiophosphate, zinc di-octyldithiophosphate, zinc di-2-ethylhexyldithiophosphate, zinc di-ndecyldithiophosphate, zinc di-n-dodecyldithiophosphate, zinc diisotridecyldithiophosphate and mixtures thereof.

[0110] The amount of the zinc dithiophosphate added in the lubricating oil is not particularly restricted. The zinc dithiophosphate is preferably contained in an amount of 0.1% or less, more preferably in an amount of 0.06% or less, most

preferably in a minimum effective amount, in terms of the phosphorus element based on the total amount of the lubricating oil in order to produce a higher friction reducing effect. When the amount of the zinc dithiophosphate exceeds 0.1%, there arises a possibility of inhibiting the excellent friction reduction effect of the oxygen-^L containing organic compound (C) at sliding surfaces of the DLC member and various metal materials, particularly iron-based material.

[0111] The zinc dithiophosphate can be prepared by any known method. For example, the zinc dithiophosphate may be prepared by reacting alcohols or phenols having the above R⁴, R⁵, R⁶ and R⁷ hydrocarbon groups with phosphorous pentasulfide to form dithiophosphoric acid, and then, neutralizing the thus-formed dithiophosphoric acid with zinc oxide. Herein, the molecular structure of zinc dithiophosphate differs according to the alcohols and the like used as a raw material for the zinc dithiophosphate production.

The other anti-friction agent or extreme-pressure additive can be exemplified by disulfides, sulfurized fats and oils, olefin sulfides, phosphate esters having one to three C2-C20 hydrocarbon groups, thiophosphate esters, phosphate esters, thiophosphite esters and amine salts of these esters.

[0112] The metallic detergent can be any metallic-detergent compound commonly used for a lubricating oil. Specific examples of the metallic detergent usable in connection with the present invention include sulfonates, phenates and salicylates of alkali metals or alkali-earth metals; and mixtures of two or more thereof. Examples of the alkali metals include sodium (Na) and potassium (K), and examples of the alkali-earth metals include calcium (Ca) and magnesium (Mg). In connection with the present invention, sodium and calcium sulfonates, sodium and calcium phenates, and sodium and calcium salicylates are suitably used. The total base number and amount of the metallic detergent can be selected in accordance with the lubricating oil performance required. The total base number of the metallic detergent is usually 0 to 500 mgKOH/g, preferably 150 to 400 mgKOH/g, as measured by perchloric acid method according to ISO 3771 "Determination of base number - Perchloric acid potentiometric titration method". The amount of the metallic detergent is usually 0.1 to 10% based on the total amount of the lubricating oil.

[0113] The antioxidant can be any antioxidant compound commonly used for a lubricating oil. Specific examples of the antioxidant usable in connection with the present invention include: phenolic antioxidants, such as 4,4-methylenebis(2,6-di-tert-butylphenol) and octadecyl-3-(3,5-di-tert-butyl-4-hydroxyphenyl)

propionate; amino antioxidants, such as phenyl- α -naphthylamine, alkylphenyl- α -naphthylamine and alkylidiphenylamine; and mixtures of two or more thereof.

The amount of the antioxidant is usually 0.01 to 5% based on the total amount of the lubricating oil.

[0114] The viscosity index improver can be concretely exemplified by: non-dispersion type viscosity index improvers, such as copolymers of one or two monomers selected from various methacrylic acids, and hydrides of the copolymers; and dispersion type viscosity index improvers, such as copolymers of methacrylates (including nitrogen compounds).

There may be also used, as the viscosity index improver, copolymers of ethylene and α -olefins (such as propylene, 1-butene and 1-pentene) and hydrides thereof, polyisobutylenes and hydrides thereof, a hydrogenated copolymer of styrene and diene, a copolymer of styrene and maleic anhydride and polyalkylstyrenes.

The molecular weight of the viscosity index improver needs to be selected in view of shear stability. For example, the number-average molecular weight of the viscosity index improver is desirably in a range of 5,000 to 1,000,000, more desirably 100,000 to 800,000, for dispersion or non-dispersion type polymethacrylates; in a range of 800 to 5,000 for polyisobutylenes and hydrides thereof; and in a range of 800 to 300,000, more desirably 10,000 to 200,000 for ethylene/ α -olefin copolymers and hydrides thereof. The above viscosity index improving compounds can be used alone or in the form of a mixture of two or more thereof. The amount of the viscosity index improver is preferably 0.1 to 40.0% based on the total amount of the lubricating oil.

[0115] The friction modifier other than the above-mentioned (C) and (D) can be exemplified by metallic friction modifier such as boric acid ester, molybdenum dithiophosphate, molybdenum dithiocarbamate, molybdenum disulfide and the like. The rust inhibitor can be exemplified by alkylbenzene sulfonates, dinonylnaphthalene sulfonates, esters of alkenylsuccinic acids and esters of polyhydric alcohols.

The nonionic surfactant and the deemulsifier can be exemplified by nonionic polyalkylene glycol surfactants, such as polyoxyethylene alkylethers, polyoxyethylene alkylphenylethers and polyoxyethylene alkyl-naphthylethers.

The metal deactivator can be exemplified by imidazoline, pyrimidine derivatives, thiadiazole, benzotriazole, thiadiazole and the like.

The anti-foaming agent can be exemplified by silicones, fluorosilicones and fluoroalkylethers.

In case that these additives are contained in the low-friction agent composition used in the present invention,

the contents of them are as follows: The friction modifier other than (C) and (D), the content of the rust inhibitor and

demulsifier can be suitably selected from the range of 0.01 to 5% based on the total amount of the composition; and the content of the metal deactivator can be suitably selected from the range of 0.0005 to 1% based on the total amount of the composition.

[0116] In the next place, a friction reduction method according to the invention will be detailed.

In such a friction reduction method, between sliding surfaces made of a DLC coated sliding member (A) covered with diamond-like carbon and a sliding member (B) that uses a metal material, a non-metal material or a coated material obtained by coating a thin film on a surface thereof, and an arbitrary combination thereof, a low-friction agent composition is interposed, at least one kind selected from a group consisting of an oxygen-containing organic compound (C) and an aliphatic amine compound (D) being supplied between the sliding surfaces. Thereby, the low friction characteristics of various sliding surfaces can be improved.

[0117]

[EXAMPLES]

In what follows, the invention will be more detailed with reference to Examples and Comparative Examples. However, the invention is not restricted to the examples.

[0118]

(Sliding member)

As an example of a sliding member, a test piece for use in a simple pinion disc friction test as shown in Fig. 1 was prepared. The simple test piece includes three pins and a disc and is prepared with a sliding member obtained according to a method described below.

- Pin material After polishing into a predetermined pin shape from a heat-treated SUJ2 material, polishing was applied with a lapping tape to finish pins into various surface roughness (Ra is 0.2 μm or less).

- Disc material

After disc-like raw materials made of a heat-treated SUJ2 material and an AC2A material were subjected to a predetermined aging process, a sliding surface with a pin was polished, followed by finishing to various surface roughness.

- Surface treatment

On surfaces of thus prepared pin materials and disc materials, by use of a PVD process or a CVD process that uses graphite as a target, materials (1) through (3) below were coated with various film thicknesses. Coated surfaces were further polished with a lapping tape and finished to various surface roughness (Ra is 0.1 μm or less).

- (1) a-C ... (PVD processing)
- (2) DLC(a-C:H) ... (CVD processing)
- (3) CrN

The sliding members are shown in Table 1.

[0119]

Table 1

Film Thickness (μm)	Surface hardness		Surface roughness Ra (μm)		Sample Oil No.	Friction Coefficient
	Disc Hv	Pin Hv	Disc	Pin		
0.5	1350	750	0.04	0.05	4	0.037
0.5	1350	750	0.04	0.05	5	0.040
1.1	1800	750	0.04	0.05	8	0.015
1.1	1800	750	0.04	0.05	9	0.04
1.1	1800	750	0.04	0.05	6	0.14

Example	Pin-on-disc material				
	Disc		Pin		
	Base material	Coating	Base material	Coating	Coating
Example 8	Heat-treated SUJ2 Material	a-C	Heat-treated SUJ2 Material		None
Example 9	Heat-treated SUJ2 Material	a-C	Heat-treated SUJ2 Material		None
Example 10	Heat-treated SUJ2 Material	a-C	Heat-treated SUJ2 Material		None
Example 11	Heat-treated SUJ2 Material	a-C	Heat-treated SUJ2 Material		None
Comparative Example 1	Heat-treated SUJ2 Material	a-C	Heat-treated SUJ2 Material		None

Table continued

Film Thick- ness	Surface hardness		Surface roughness Ra (μm)		Sample Oil No.	Friction Coefficient
	Disc Hv	Pin Hv	Disc	Pin		
	1800	750	0.04	0.05	7	0.098
1.1	1700	750	0.04	0.03	4	0.132
1.1	1700	750	0.04	0.03	2	0.148
1.1	1700	750	0.04	0.03	7	0.056

Example	Pin-on-disc material					Disc
	Disc		Pin			
	Base material	Coating	Base material	Coating		
Comparative Example 2	Heat-treated SUJ2 Material	a-C	Heat-treated SUJ2 Material	None		1.1
Comparative Example 3	Heat-treated SUJ2 Material	PVD/CrN	Heat-treated SUJ2 Material	None		2.0
Comparative Example 4	Heat-treated SUJ2 Material	PVD/CrN	Heat-treated SUJ2 Material	None		2.0
Reference Example 1	Heat-treated SUJ2 Material	PVD/CrN	Heat-treated SUJ2 Material	None		2.0

[0120]

(Preparation of low-^L friction agent composition)

- Sample Oil 1

As lubricating oil base oil, hydrogenated 1-decene oligomer (kinematic viscosity at 100°C: 3.9 mm²/s, viscosity index: 124 and total aromatic content: 0.0%) was used. Thereto, 1.0% of oleyl alcohol as an oxygen-containing organic compound, 13.0% of other additive (5.0% of ash-^L less dispersant: polybutenyl succinic imide (nitrogen content: 1.2%), metal base detergent: 0.5% of calcium sulfonate (total base number: 300 mgKOH/g and calcium content: 12.0%) and 0.9% of calcium phenate (total base number: 255 mgKOH/g and calcium content: 9.2%), a viscosity index improver, an oxidation inhibitor, an extreme pressure agent, a rust-preventive, an anti-emulsifying agent, a nonionic surfactant, a metal deactivator, a defoaming agent and the like) were blended to prepare.

- Sample Oil 2

Except that 13.0% of other additive was not added, an operation same as sample 1 was repeated to prepare.

- Sample Oil 3

Except that oleic acid was used as an oxygen-containing organic compound, an operation same as sample 1 was repeated to prepare.

- Sample Oil 4

Except that 0.5% of oleyl alcohol and 0.5% of oleic acid were used as the oxygen-containing organic compound, an operation same as sample 1 was repeated to prepare.

- Sample Oil 5

Except that with hydrocracked mineral oil (kinetic viscosity at 100°C: 5.0 mm²/s, the viscosity index: 120 and total aromatic content: 5.5%) as lubricating oil base oil, 1.0% of oleic acid amide was added as an oxygen-containing organic compound, an operation same as sample 1 was repeated to prepare.

- Sample Oil 6

Commercially available engine oil of which kinetic viscosity at 100°C is 10.2 mm²/s was used.

- Sample Oil 7

Except that in place of oleic acid amide 1.0% of molybdenum dithiocarbamate was added, an operation same as sample 5 was repeated to prepare.

- Sample Oil 8

Glycerin (Glycol Anhydrous: 1,2,3-Propanetriol, Fluka, manufactured by Sigma Aldrich Japan Co.,) was used singularly (100%).

- Sample Oil 9

A tri-ester (KAOLUBE KSL-268) of trimethylol propane and a mixture of aliphatic acids i-C8, n-C8, C10 and C11 was used singularly (100%).

[0121]

Table 2

Table 2

Sample oil 11										100		Unmeasured	1) PAO (poly α -olefin) (kinematic viscosity at 100°C: 3.9 mm ² /s,
Sample oil 10		100									13.0	10.2	
Sample oil 9								100				5.3	
Sample oil 8							100					Unmeasured	

	Sample oil 1	Sample oil 2	Sample oil 3	Sample oil 4	Sample oil 5	Sample oil 6 ⁹⁾	Sample oil 7
Synthetic Oil ¹⁾	100	100	100	100	-	Unmeasured	-
Mineral oil ²⁾	-	-	-	-	100		100
Oleyl alcohol ³⁾	1.00	1.00	-	0.50	-		-
Oleic acid ⁴⁾	-	-	1.00	0.50	-		-
Oleyl amide ⁵⁾	-	-	-	-	1.00		-
Molybdenum compound ⁶⁾	-	-	-	-	-		1.00
Glycerin ⁹⁾	-	-	-	-	-		-
Trimethylolpropane ester	-	-	-	-	-		-
Glycerol monooleyl ether	-	-	-	-	-		-
Glycerol mono-2-ethylhexyl ether	-	-	-	-	-		-
Other additive ⁷⁾	13.0		13.0	13.0	13.0		13.0
Sample oil characteristics: Kinematic viscosity (100°C)	10.2	3.9	10.2	10.3	10.3	10.2	10.3

- 3) Oleyl alcohol
- 4) Oleic acid
- 5) Oleic acid amide
- 6) Molybdenum dithiocarbamate (molybdenum content: 9.9% by mass)
- 7) As other additives, ash-less dispersing agent, a viscosity index improver, an oxidation inhibitor, an extreme pressure agent, a rust-preventive, an anti-emulsifying agent, a nonionic surfactant, a metal deactivator and a defoaming agent are included.
- 8) Commercially available SG oil
- 9) Glycerin (Fluka 49707 Glycerin anhydrous: 1,2,3-Propanetriol)
- 10) Trimethylol propane (trimethylol propane iso-C8, n-C8, C10, 11 ester)

[0122]

(Examples 1 through 11)

As shown in Table 1, the respective sliding members were combined, further with the respective low-^L friction agent compositions (the sample oils 1 through 5 and 8 and 9) described together in Table 1, low-friction sliding mechanisms were prepared, followed by carrying out a pin-on-disc friction test described below. Results thereof are shown together in Table 1.

[0123]

[Pin-on-disc Friction Test]

Pressure at maximum hertz: 80 MPa

Disc rotation speed: 30 rpm

Oil supply method: oil bath

Temperature of supplied oil: 80°C

Test time period: 60 min

[0124]

(Comparative Examples 1 through 4)

Similarly to examples, the respective sliding members were combined, further with the respective low-friction agent compositions (the sample oils 1, 2, 6 and 7) described together in Table 1, low-friction sliding mechanisms were prepared, followed by carrying out a pin-on-disc friction test. Results thereof are shown together in Table 1.

[0125] From Table 1, it is found that all combinations of the base materials and the sample oils obtained in Examples 1 through 11 show excellent low friction coefficients. For instance, these are, in comparison with Comparative Example 1 where a combination of a base material that is used in a general gasoline engine and sample oil 6 is used, could obtain the friction reduction effect of substantially 50 to 70%.

Furthermore, from results of Examples 6 through 8, it is found that a hydroxyl group in the low-friction agent composition is excellent in the friction reduction effect.

Still furthermore, from results of Examples 10 and 11, it is found that a compound having an alcoholic hydroxyl group is excellent in the friction reduction effect.

Test pieces obtained in Examples 1 through 11 each did not have any problem in a surface shape after the test, were very excellent in the wear resistance as well, and showed stable low friction characteristics.

[0126] A combination of a test piece base material and a low-friction agent composition according to Reference Example 1 is a combination where steel materials one of which is CrN-treated one and organic molybdenum that is said to be the most effective as the low-friction agent composition for sliding surfaces between steel materials are combined. As a result, the friction coefficient shows such a low value as substantially 0.06. This is inferred that similarly to a mechanism so far studied in various fields, a molybdenum disulfide film is formed on a sliding surface and thereby low friction is obtained. In the invention, the low friction is a target, and it is found that the low friction characteristics in the foregoing examples are very excellent.

[0127] Furthermore, a combination of the test piece base material and the low-friction agent composition according to Comparative Example 1 is a combination of general bearing steel and engine oil. As a result, the friction coefficient exceeds 0.1 to be poor in the friction characteristics. This can be assumed that the mechanism showing a low friction behavior suggested in the invention did not sufficiently work.

On the other hand, Comparative Example 2 is a combination where steel materials one of which is DLC-treated and sample oil blended with an organic molybdenum compound are combined. The friction coefficient was such high as substantially 0.1, that is, an advantage such as that of Example 2 was not obtained. This is inferred that the friction reduction mechanism is different from that of a sliding surface between existing steel materials.

Furthermore, Comparative Examples 3 and 4 are a combination where steel material obtained by applying a CrN treatment on a base material and used in Comparative Example 2 and sample oil used in the examples, that is, sample oil that can obtain the low friction effect when applied to steel materials one of which is DLC-treated are combined. As a result, the friction coefficient showed such a large value as exceeding 0.1. It is assumed that since this is a different combination from that of a metal material and sample oil shown in the invention, the lower friction cannot be achieved.

[0128] A combination of a test piece base material and a low-friction agent composition according to Reference Example 1 is a combination where steel materials one of which is CrN-treated and sample oil 7 where organic molybdenum that was most effective as the low-^L friction agent composition for sliding surfaces between existing steel materials was blended are combined. As a result, the friction coefficient shows such a low value as substantially 0.05. This

is inferred that similarly to a mechanism so far studied in various fields, a molybdenum disulfide film was formed on a sliding surface and thereby low friction was obtained.

[0129] In the above, Examples and Comparative Examples according to the invention are more detailed; however, the invention is not restricted thereto, and, as far as it is within the gist of the invention, various modifications can be applied.

For instance, the invention can be applied to gear sliding members used in industrial machinery as well.

[0130]

[EFFECT OF THE INVENTION]

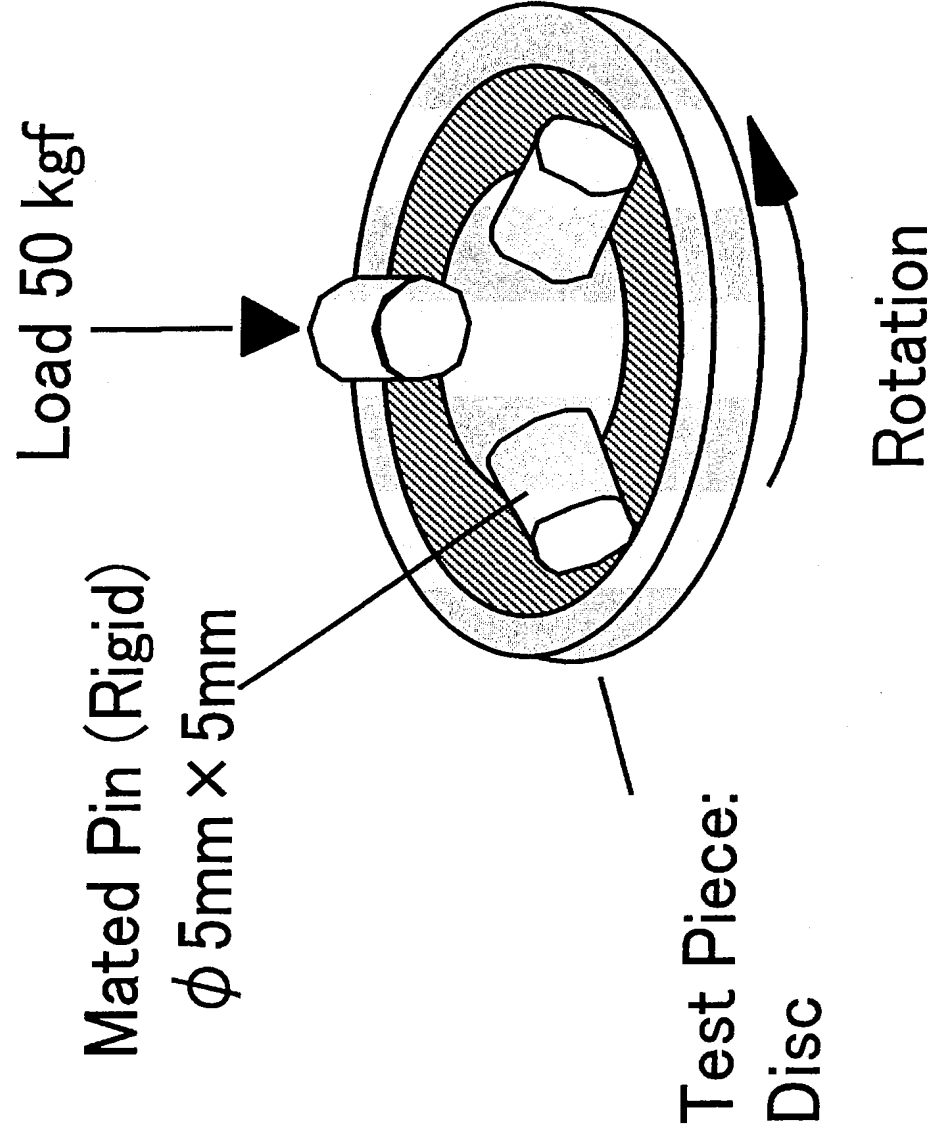
As described above, according to the present invention, since at least one adopts a combination of a sliding member as a DCL material and a specific compound, there can be provided a low-friction sliding mechanism, a low-^L friction agent composition and a friction reduction method that can exert very excellent low friction characteristics to a sliding surface present under various applications, and, in particular, that have more excellent low friction characteristics than that of a combination of the existing steel material and the organic Mo compound.

[BRIEF DESCRIPTION OF DRAWINGS]

Fig. 1 is a schematic diagram showing an example of a pin-on-disc friction test.

[DOCUMENT TITLE] DRAWING

Figure 1



[DOCUMENT TITLE] ABSTRACT

[ABSTRACT]

[PROBLEM] To provided a low-friction sliding mechanism, a low-friction agent composition and a friction reduction method that can exert very excellent low friction characteristics to a sliding surface present under various applications, and, in particular, that have more excellent low friction characteristics than that of a combination of the existing steel material and the organic Mo compound.

[MEANS FOR THE SOLUTION] 1. A low-friction sliding mechanism wherein an oxygen-containing organic compound (C) or a low-friction agent composition (D) is interposed between sliding surfaces of a DLC coated sliding member (A) and a sliding member (B), characterized in that the DLC coated sliding member (A) is formed by coating diamond-like carbon on a base material; the sliding member (B) is formed with at least one kind of material selected from a group consisting of a metal material, a non-metal material and a coated material obtained by coating a thin film on a surface of the metal material or the non-metal material; the oxygen-containing organic compound (C) includes an oxygen in a molecule; and the low-friction agent composition (D) contains the oxygen-containing organic compound (C).

[SELECTIVE DRWAING] NONE